Science Opportunities using NSLS II: Fast timing and "Speckle" Experiments

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Outline

Two types of time resolved x-ray scattering experiments:

- **"Classical" experiments**
  - **Previous examples**
  - **Possible futures**
- **"Speckle" experiments**
  - **Example of wide-angle speckle experiment**
  - **Possible futures**
- **Discussion of NSLS II**

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Motivation for Ultrafast Structural Studies

Our fundamental understanding of the static or time-averaged structure of matter on atomic length scales has been dramatically advanced by direct structural measurements using x-rays.

Example: Atomic positions in core of Reovirus

- molecular mass of 52 million
- unit cell of 1255Å
- core is 700Å in diameter
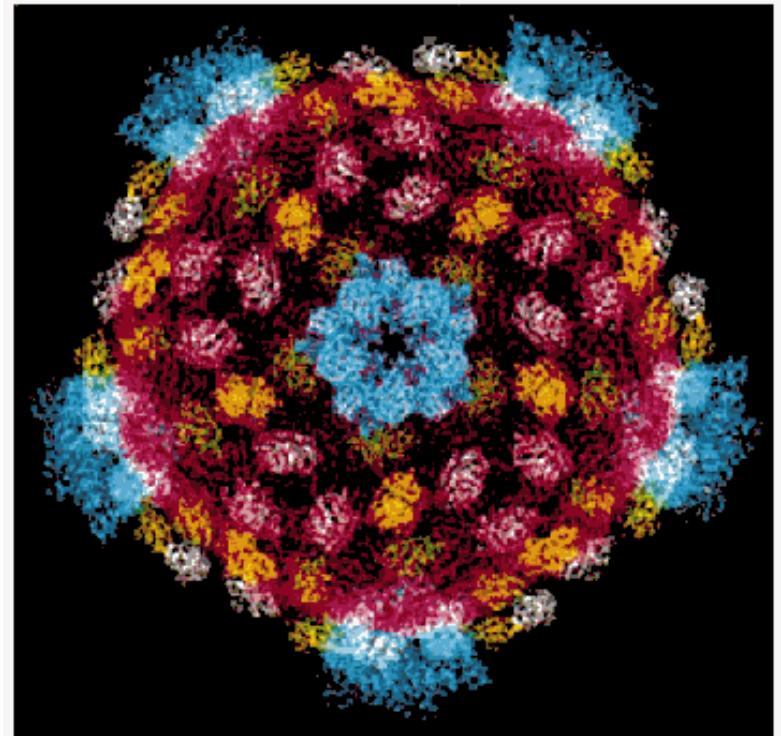
K.M. Reinish, M.L. Nibert & S. C. Harrison, Cell, **100**, 345-356 (2000).

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Motivation for Ultrafast Structural Studies
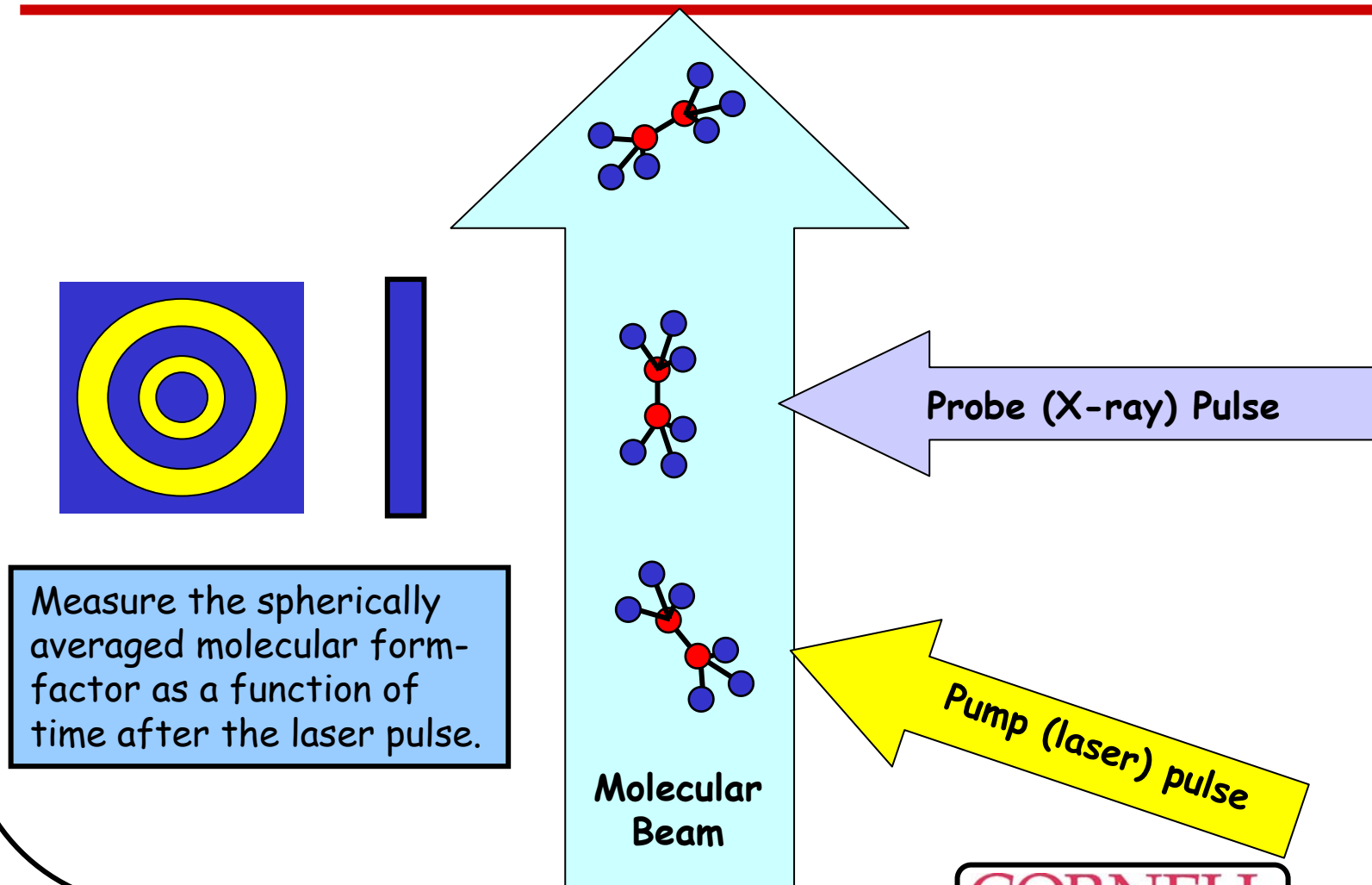
However, the structure of matter is not static.

Developing our understanding the fundamental behavior of matter requires structural measurements on the time-scale on which atoms rearrange.

Since the evolution of atomic-scale structure is determined by making and breaking chemical bonds and rearranging atoms, the evolution occurs on the time scale of a vibrational period, ~100 fs.

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Pump/Probe Chemistry

Measure the spherically averaged molecular form-factor as a function of time after the laser pulse.

Probe (X-ray) Pulse

Pump (laser) pulse

Molecular Beam

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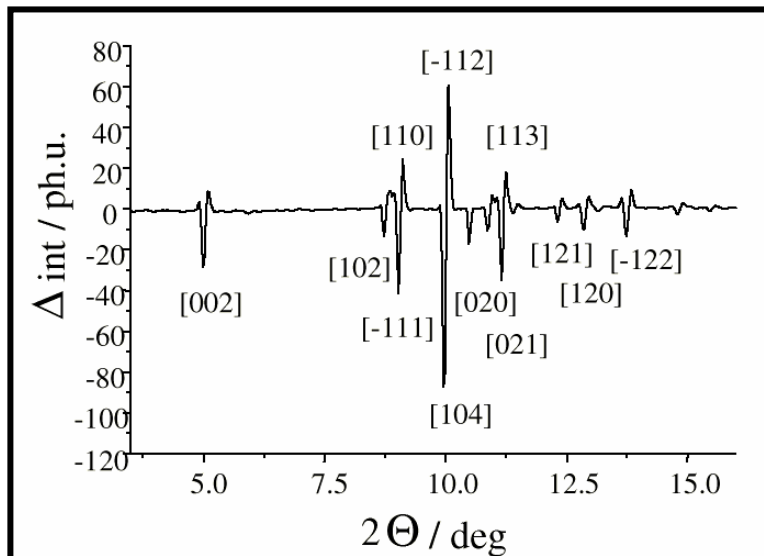
Ultrafast Chemical Reactions

Scientific challenge is to understand the structural evolution of the "transition state(s)" intermediate between reactant and product species.
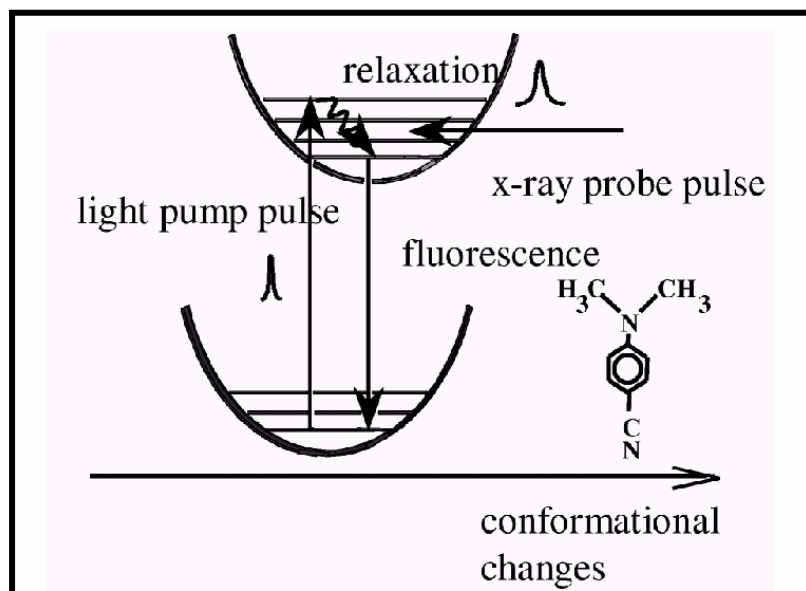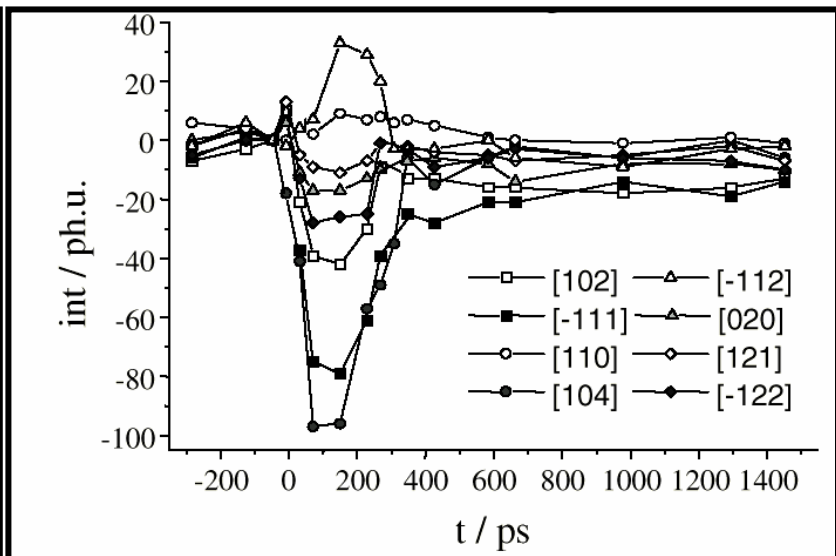

S. Techert, F. Schotte, and M. Wulff, Phys. Rev. Lett. **86**, 2030-2033 (2001).

Picosecond Crystallography

Photo-excitation of $C_9H_{10}N_2$

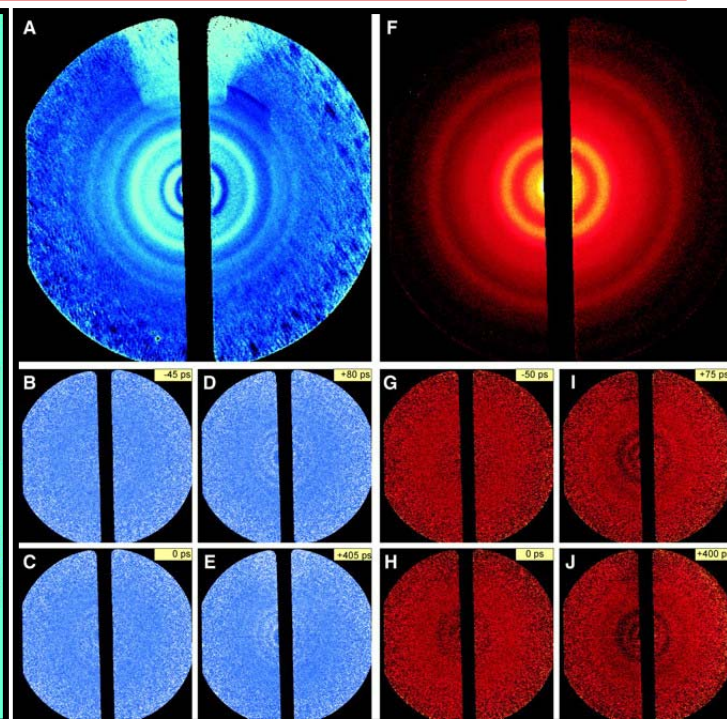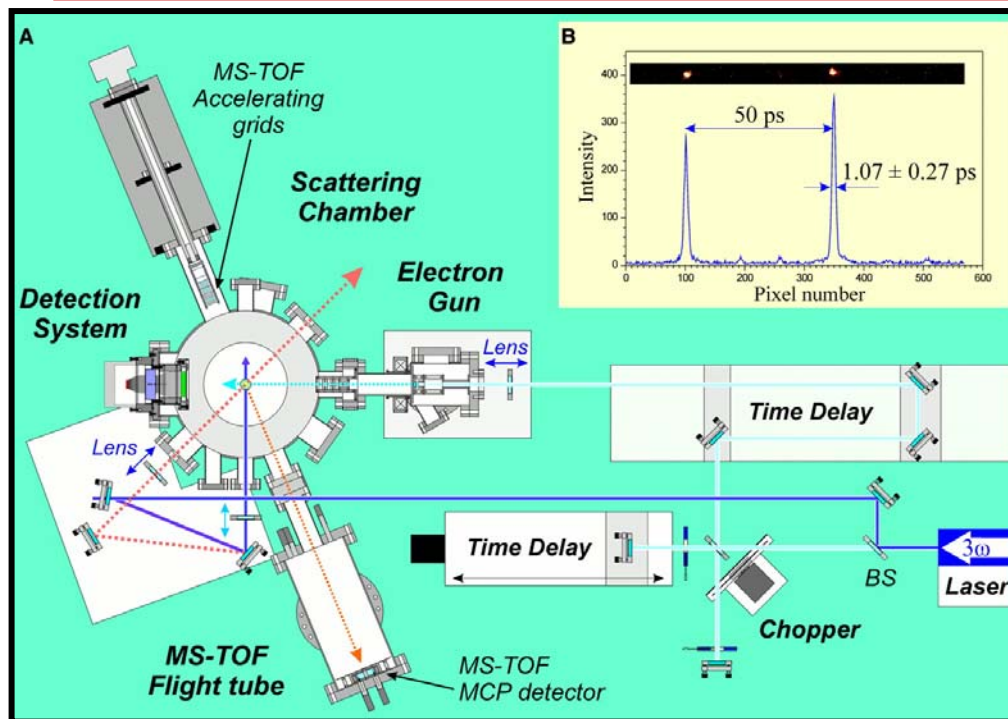Intensity difference map of powder pattern @ 80 ps.

Change of the integrated Bragg intensities as a function of time.

S. Techert, F. Schotte, and M. Wulff, Phys. Rev. Lett. **86**, 2030-2033 (2001).

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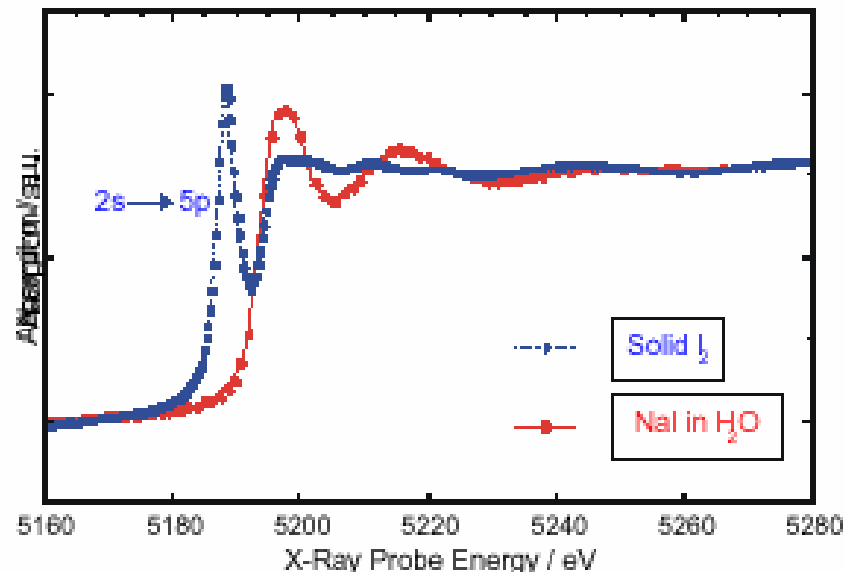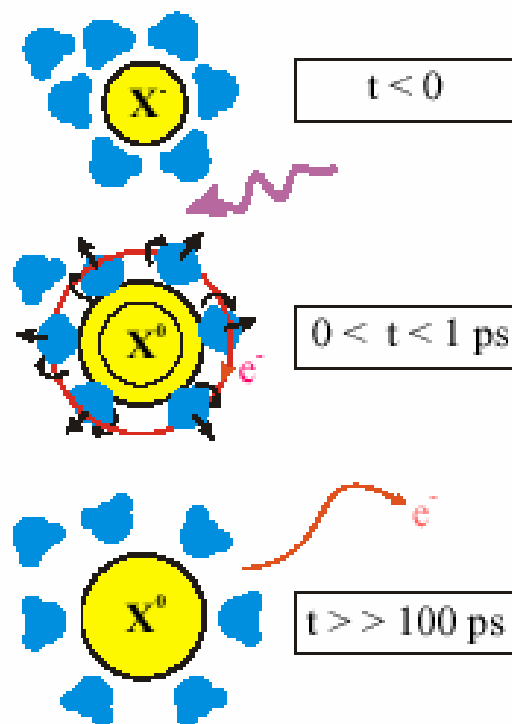
Ultrafast Structure Studies

Caltech's Ultrafast Electron Diffraction (UED) apparatus.

UED images of $C_2F_4I_2$ (blue) and 1,3-cyclohexadiene (red).

H. Ihee, V.A. Lobastov, U.M. Gomez, B.M. Goodson, R. Srinivasan, C.-Y. Ruan, and A. Zewail, Science **291**, 458-462 (2001).

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Hydration Dynamics

Schematic illustration of Photo-neutralization of I- in liquid phase. EXAFS of 2s → 5p. Change in spectra arises from changed I-O distances. (From Schoenlein & Falcone)
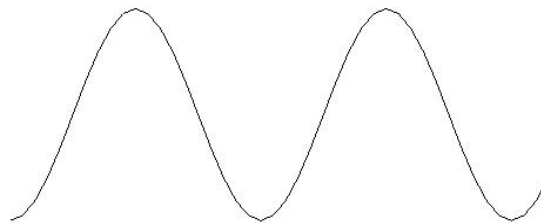
Electronic Systems

Charge Density Waves (CDWs) are a prototypic system for driven correlated systems with large numbers of degrees of freedom in the presence of random disorder.

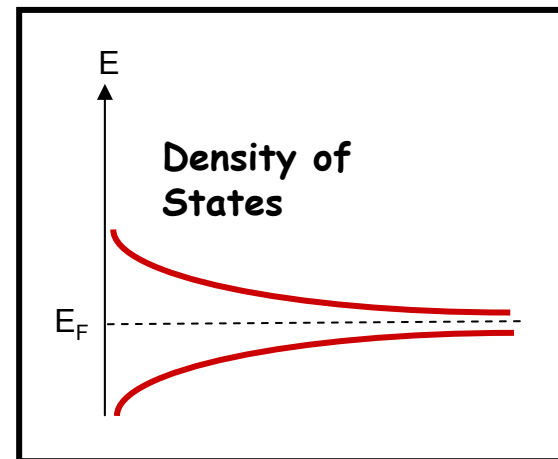$NbSe_3$ is one of the best characterized systems that is frequently used as a reference point for other CDW systems.

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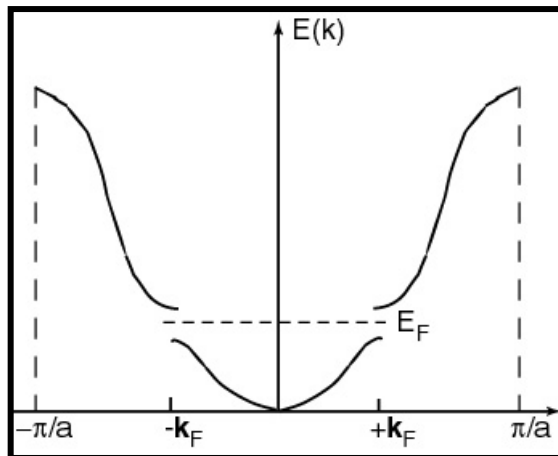UNIVERSITY

Diffraction from CDWs

Conduction electron density

Ionic Cores

E(k)

$-\pi/a$ $-k_F$ $+k_F$ π/a

E_F

E

Density of States

E_F

NbSe$_3$ Properties

- monoclinic unit cell
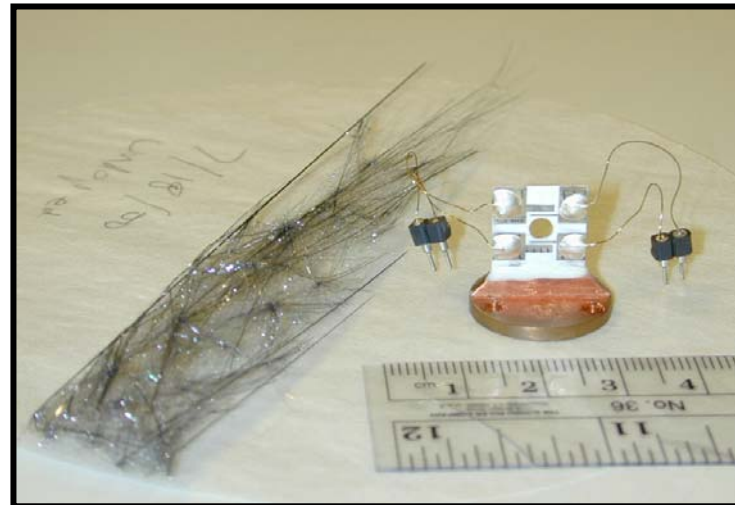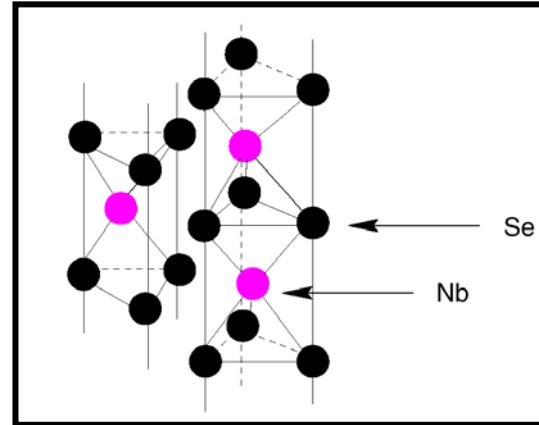
$a = 10.009 \, \AA$
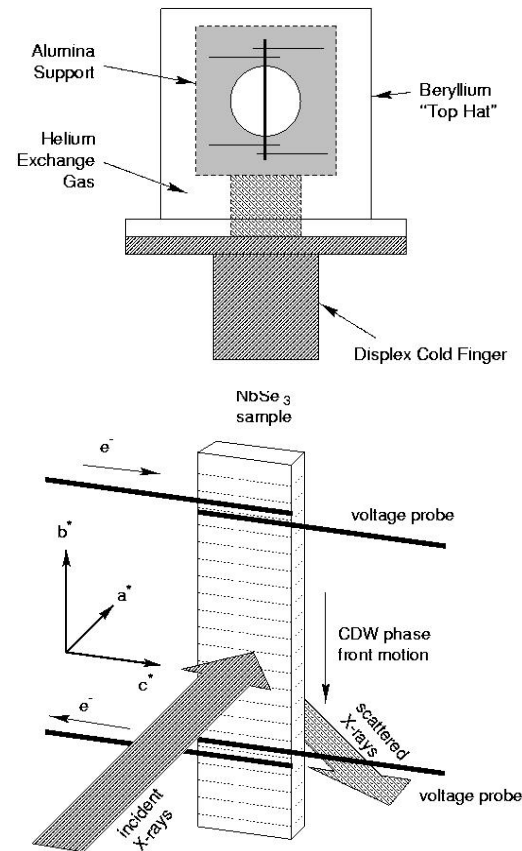
$b = 3.4805 \, \AA$ $\beta = 109.47$

$c = 15.629 \, \AA$

Hodeau *et al.*, J. Phys. C **11**, 4117 (1978)

- whisker axis || to **b, b***

- width || **c**

- thickness || **a***

- $T_{P1} \approx 145$ K, $T_{P2} \approx 59$ K

- $Q_1 \approx (0 \; 0.243 \; 0)$

- $Q_2 \approx (0.5 \; 0.26 \; 0.5)$

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Experimental Geometry

- **Beryllium Top Hat**
  - **x-ray transparent**
  - **contains He exchange gas**
- **Alumina Substrate**
  - **supports NbSe$_3$ whisker**
  - **four point connections**
- **Transmission Scattering Geometry**

Typical Pinning Data

- Current is on for $t < 0$
- Current is off for $t > 0$
- CDW satellite responds by
  - sharpening
  - (possibly) small rotation
  - integrated intensity remains constant
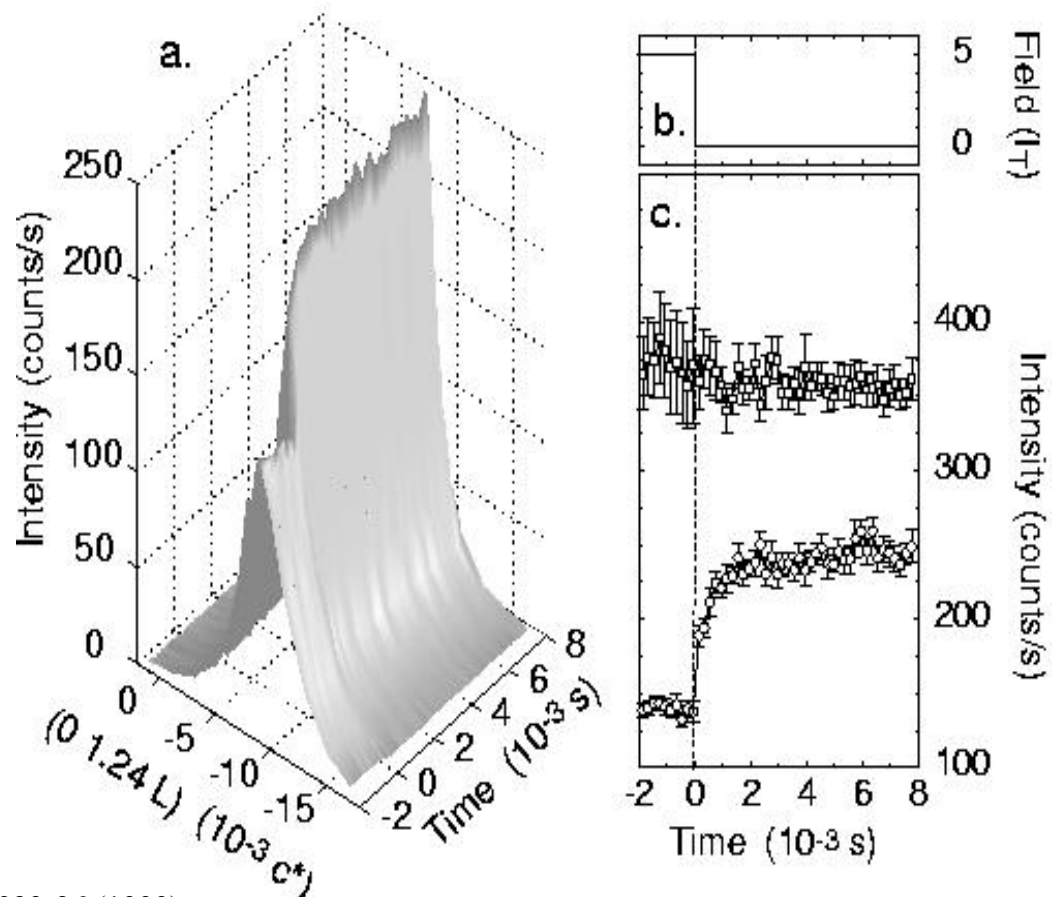
K.L. Ringland *et al.*, PRL **82**(9), 1923-26 (1999).
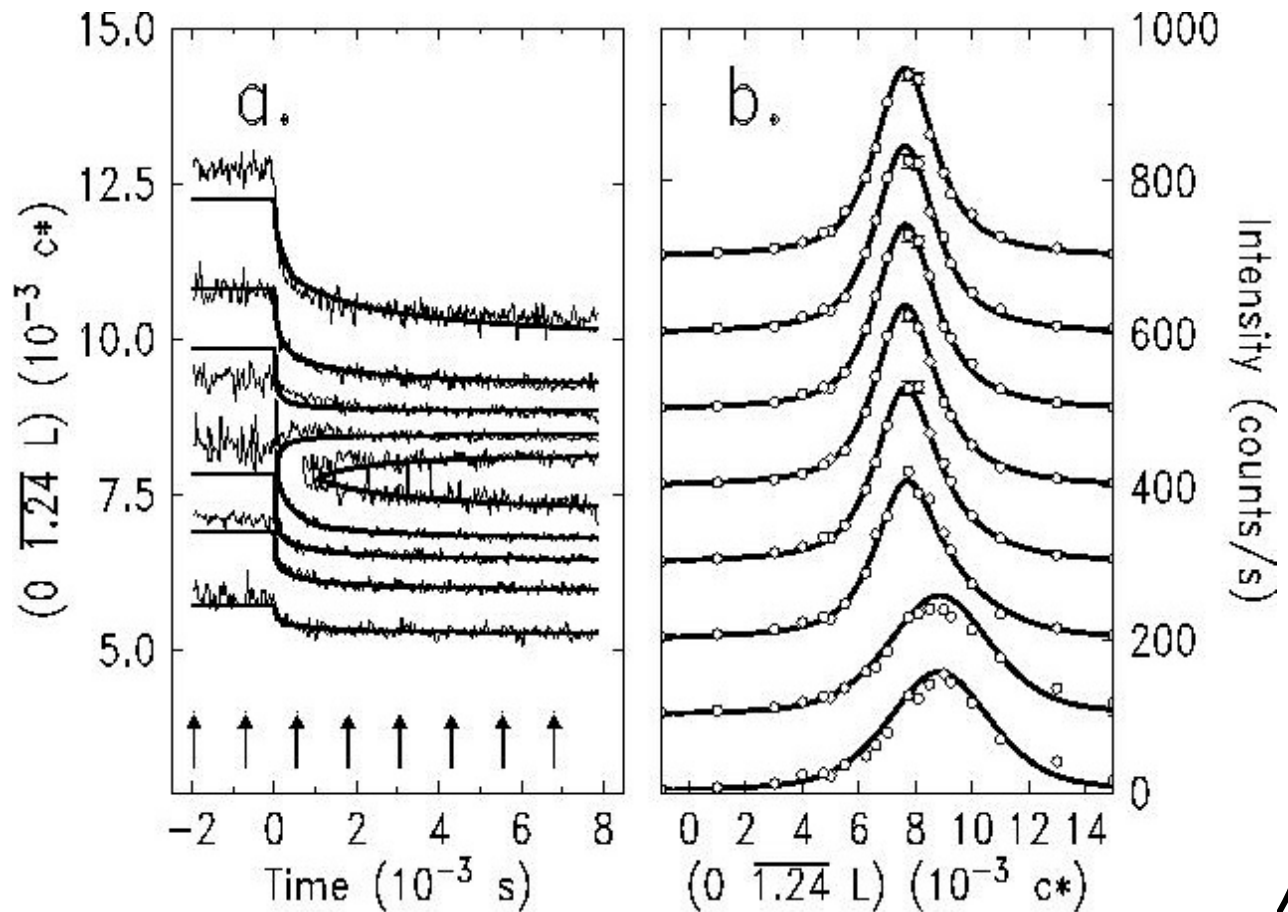
Approximate form for S(q,t)

$$S(q_\perp, t) \approx \frac{\xi_f}{1 + \xi_f^2 (q_\perp - G_\perp)^2} \left[1 - e^{-(t/\tau)^\mu} \right] \quad + \quad S(q_\perp, t=0)\, e^{-(t/\tau)^\mu}$$

Simple "stretched exponential" kinetics

Results of Analysis

- Solid lines are best fit results using only data from $t > \tau$
- 2 parameter fit (μ and τ)
- $\chi^2 \approx 1.5$
- $\sim 2 \times 10^4$ degrees of freedom.
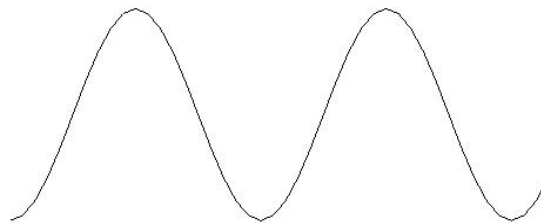- Arrows indicate times for the "slices" depicted.

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Results

- **Elastic (FLR) theory works for pinned state structure.**
- **Elastic theory gives correct line-shape above threshold but incorrect numerical value for exponent.**
- **Time resolved measurements demonstrate pinning dynamics are not correctly described by elastic theory.**
- **X-rays do not observe any structural signal associated with dramatic transport phenomena (e.g., mode-locking).**

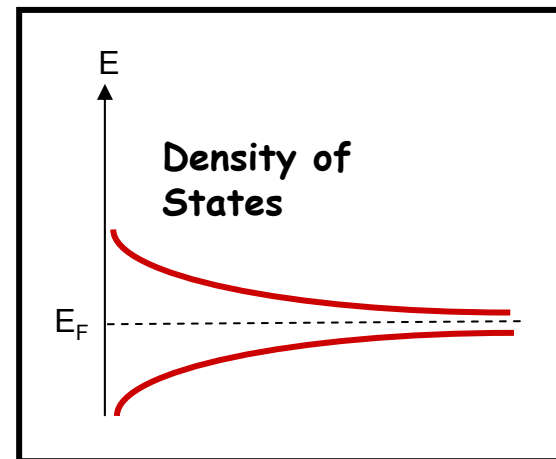**Suggests that lattice distortion wave is decoupling from conduction electron density wave**
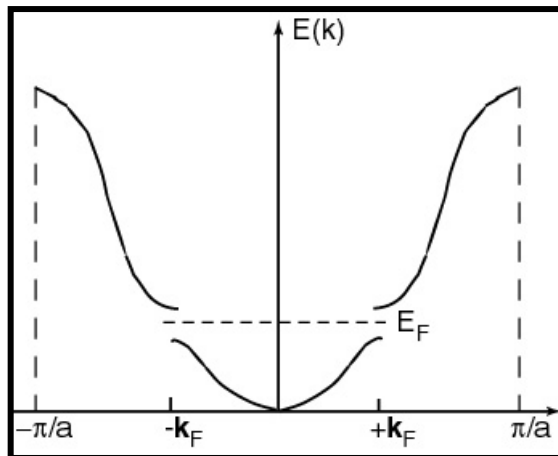
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Ultrafast Diffraction from CDWs

Conduction
electron density

Ionic Cores

$E(k)$

E_F

$-\pi/a$ $-k_F$ $+k_F$ π/a

E

Density of States
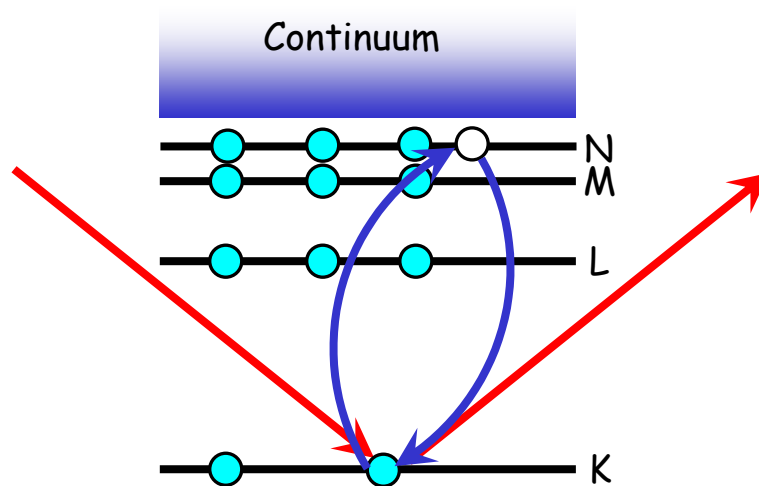
E_F

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Ultrafast Diffraction

Example: Charge-Density Waves

Resonant Scattering

Continuum

N
M

L

K

Need diffraction to amplify signal. But, diffraction is from a periodic structure.

• Electrons respond on time-scales determined by Fermi velocity.

• Lattice responds on time-scales determined by the speed of sound.

Use resonant x-ray scattering to sample unoccupied density of states of conduction band.
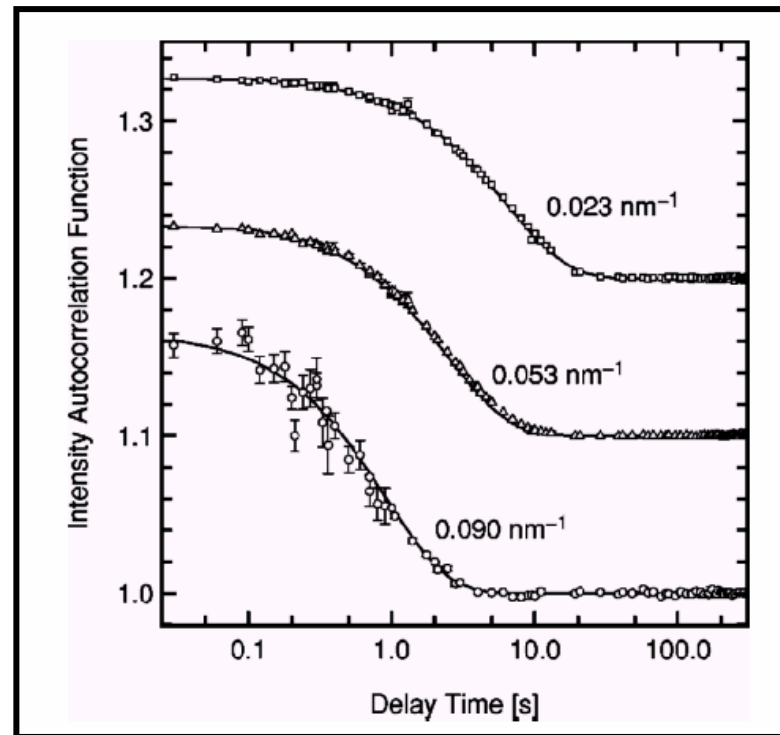
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XPCS or "Speckle" Experiments
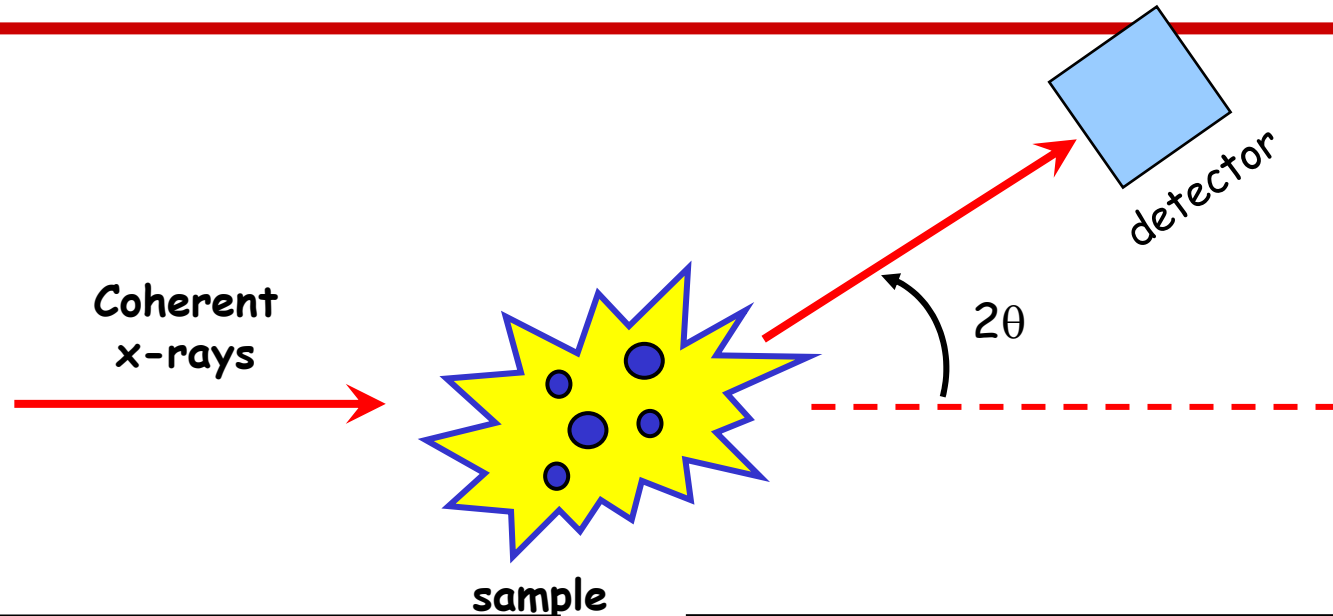
$$g_2(\mathbf{Q},t) = \frac{\langle I(\mathbf{Q},t')I(\mathbf{Q},t'+t)\rangle}{\langle I(\mathbf{Q},t')\rangle^2};$$

$$g_2(\mathbf{s}_1 - \mathbf{s}_2, t_1 - t_2) = \frac{\langle |E_s(t_1)|^2 |E_s(t_2)|^2\rangle}{\langle |E_s|^2\rangle^2}$$

Allows measurement of very "slow" dynamics at finite q in opaque materials.

Colloidal Suspension

A dense system of hard-sphere colloidal polystyrene particles, with a nominal radius of 71 nm, suspended in glycerol at a volume fraction of $\phi = 50.28$.

D. Lumma, L.B. Lurio, S.G.J. Mochrie, and M. Sutton, Rev. Sci. Instrum., **71**(9), 3274-3289 (2000).

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Limits

Transverse coherence length.

$$\xi = \lambda R' / (2\pi\sigma)$$

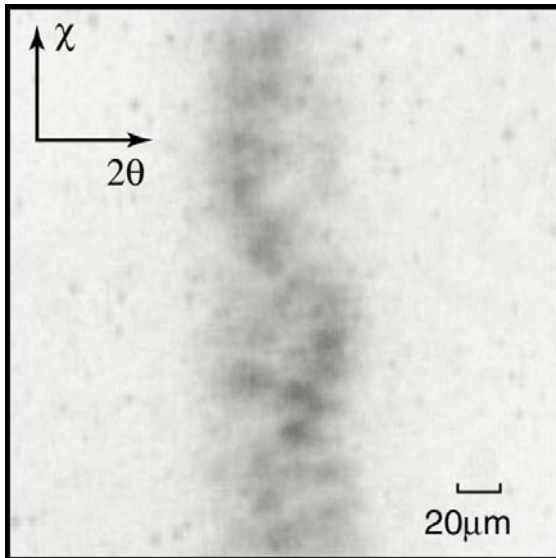State of the Art: IMM-CAT (sector 8) at APS (σ values)

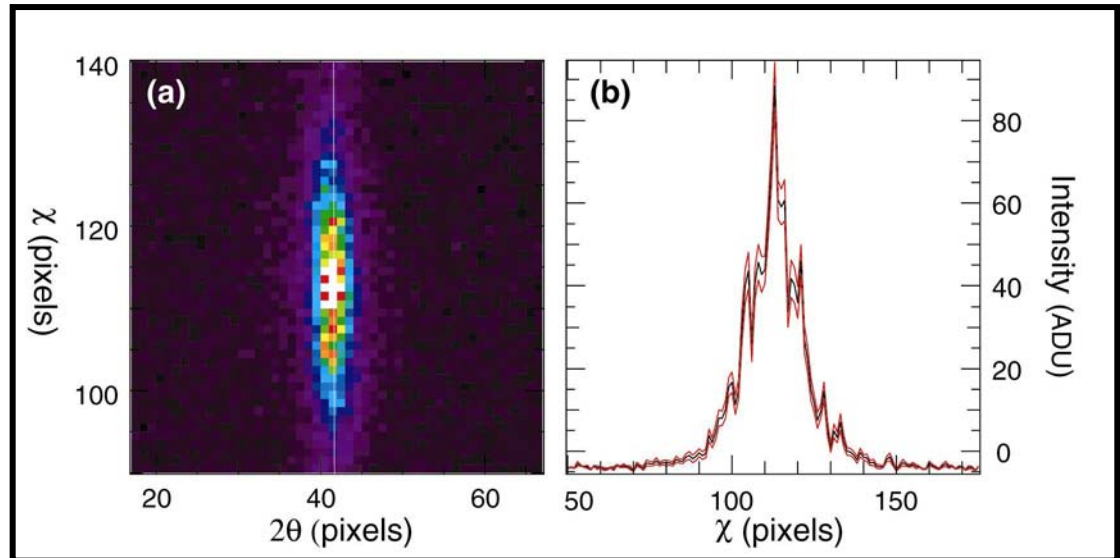$$\xi_{hor} \approx 4\mu m$$
$$\xi_{vert} \approx 28\mu m$$

Restricted to using whiskers with widths less than 10 μm.

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Coherent X-rays and Speckle: Equilibrium Dynamics

High resolution photograph of (0 1+q 0) CDW satellite in NbSe$_3$.

(a) False color CCD image of (0 1+q 0) CDW satellite in NbSe$_3$. (b) Slice of image shown in (a).

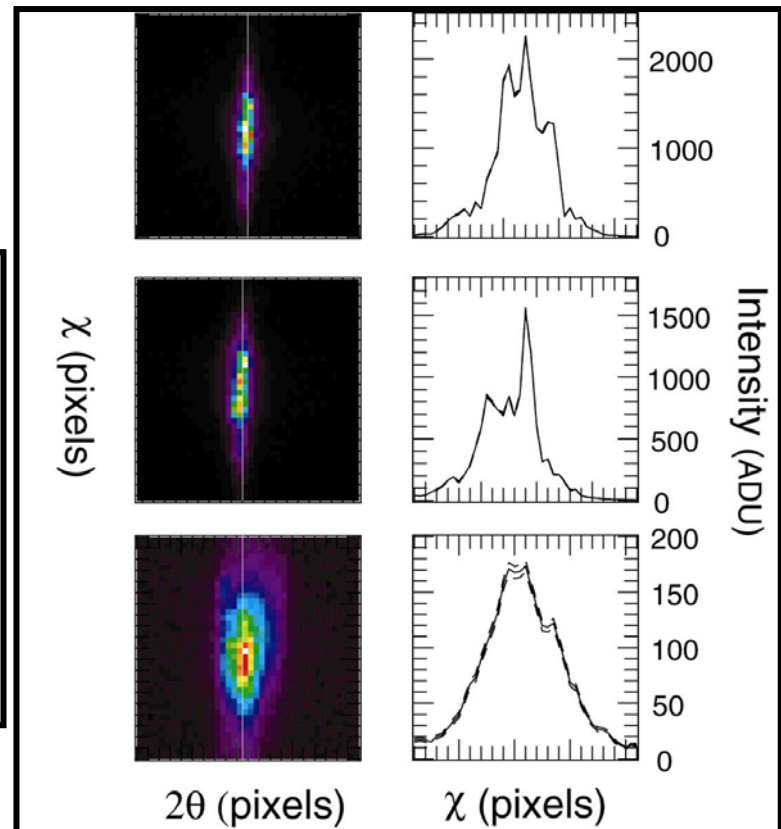Y. Li, R.E. Thorne, M. Sutton, and J.D. Brock, unpublished data.

Field Broadening

Q$_1$ CDW, T = 100K

Top: I = 0

Center: I = 0.8 × I$_T$

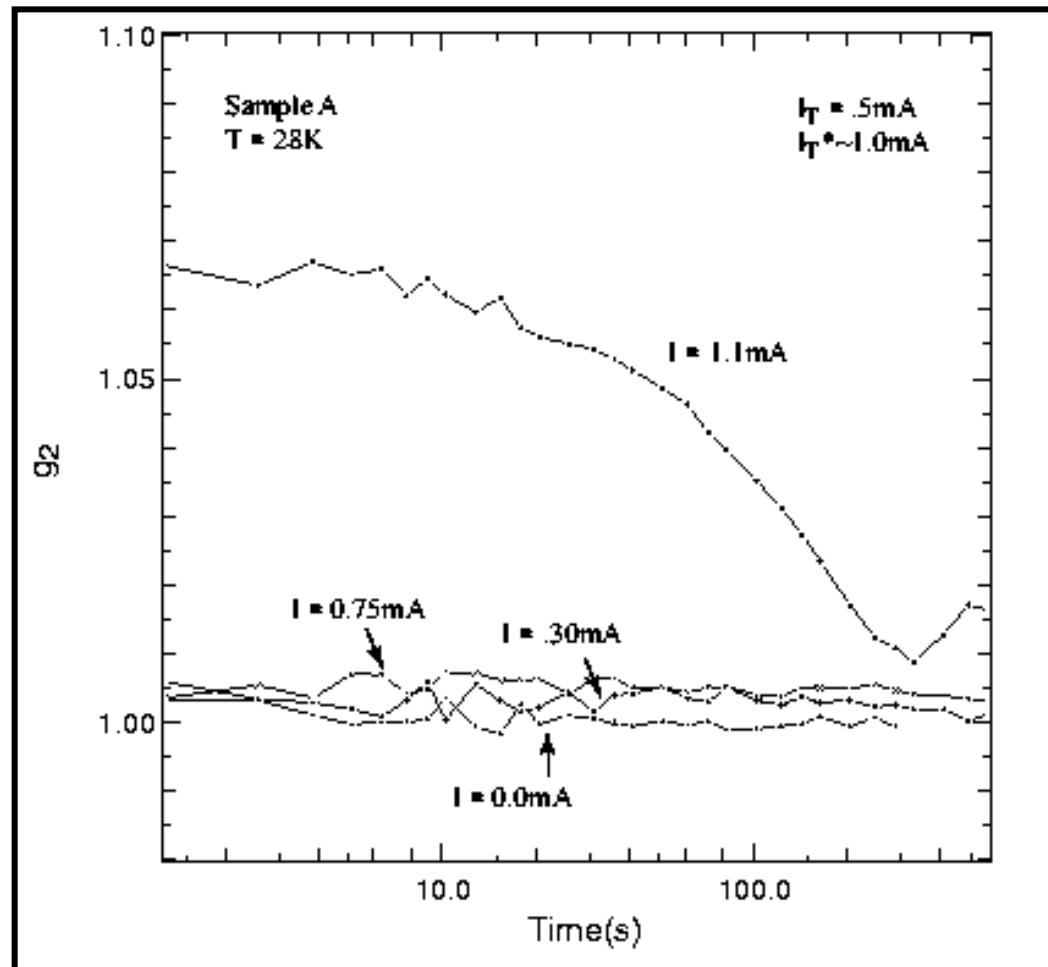Bottom: I = 2 × I$_T$

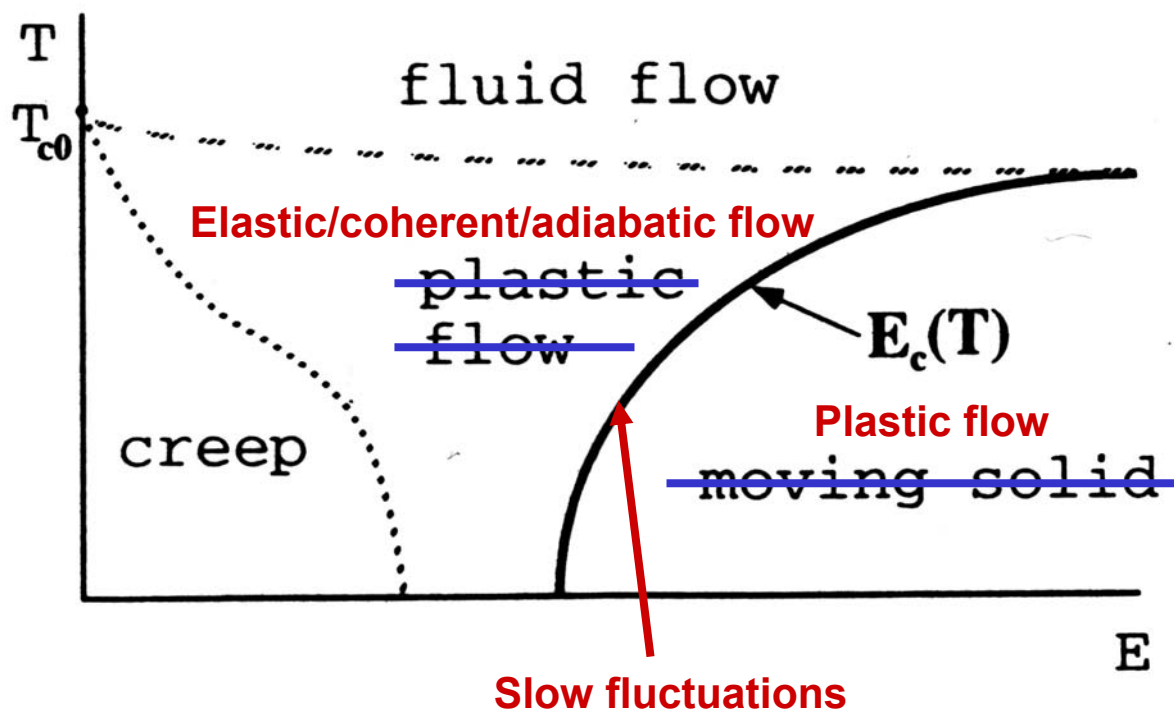Averaged over 200 frames and 5 sec each.

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Autocorrelation in Time

$E < E_T$:   Static
$E \geq E_T$:   Static
$E \approx E_T^*$:   Dynamic
$E \gg E_T^*$: "Fast"
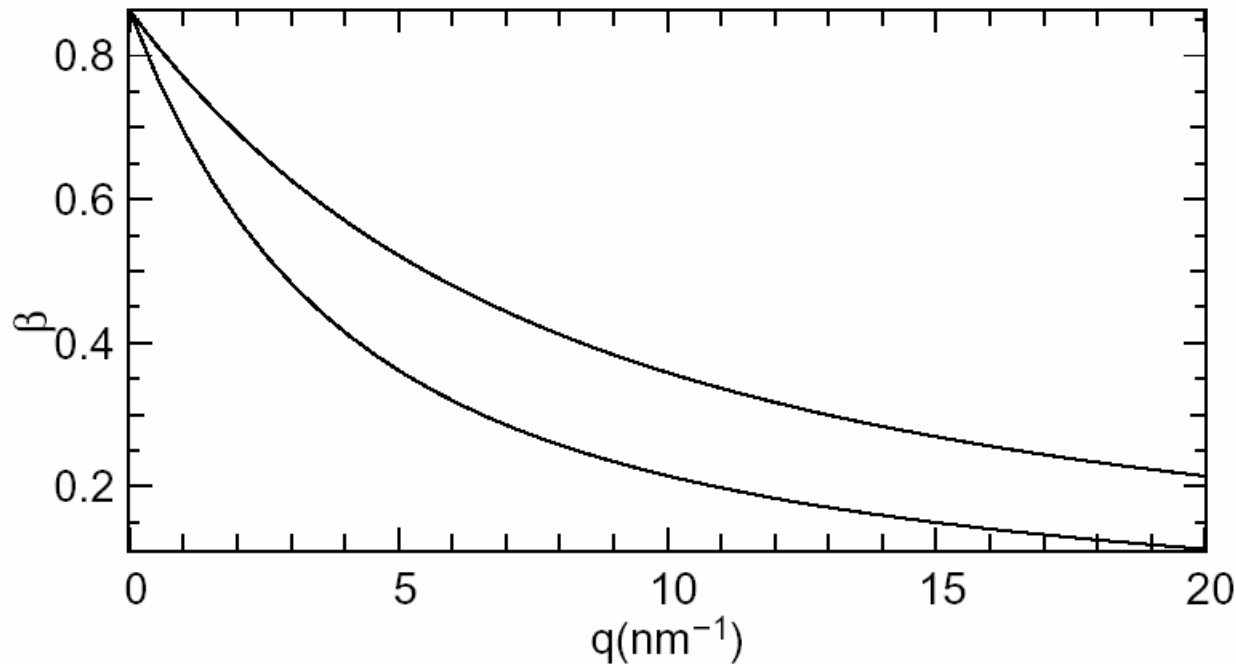
Summary

WAXS: Effect of q on coherence

β versus wavevector calculated for APS undulator A. A slit of 4×10 μm (H×V) is 55 m from the source, the energy is 7.66 KeV with $\Delta E/E = 7 \times 10^{-5}$ (C(111)) and the sample is 5 μm thick. The upper line is for reflection geometry and the lower line for transmission.
(Courtesy Mark Sutton)

Comparative Parameters

	APS (Und A)	ESRF	NSLS II
Average Brightness [photons/sec/ 0.1%bw/mm^2/ mrad^2]	1.5×10^{19}	3.1×10^{20}	$10^{20} - 10^{21}$
Average flux [photons/sec/ 0.1%bw]	7×10^{14}		$10^{15} - 10^{16}$
Pulse Length [psec]	73	35	13

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Charge

Question: "Specifically, what would an extra factor of 10 in coherent flux over present APS buy you in these experiments? What could you then do in the field of strongly correlated electron systems?"

Impact of NSLS II

(1) Increased coherent flux
allows experiments to probe

- faster times

- higher q's

- more weakly scattering samples

Coherent Flux ~ B $(\lambda^2)(\delta\lambda/\lambda)$

(2) Need increased brilliance

At small q

    contrast ~ (coherence volume) / (scattering volume)

At high q

    contrast decreases as path length increases

(3) Need full transverse (and increased longitudinal) coherence.

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Conclusions

Exciting scientific opportunities driven by increased brilliance.

- Ultra-fast structural studies

- XPCS (Speckle) Experiments

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